

Build up of off-diagonal long-range order in microcavity exciton-polaritons across the parametric threshold

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We report an experimental study of the spontaneous spatial and temporal coherence of polariton condensates generated in the optical parametric oscillator configuration, below and at the parametric threshold, and as a function of condensate area. Above the threshold we obtain very long coherence times (up to 3 ns) and a spatial coherence extending over the entire condensate (40 μm). The very long coherence time and its dependence on condensate area and pump power reflect the suppression of polariton-polariton interactions by an effect equivalent to motional narrowing.

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Planar microcavity exciton-polaritons are composite particles generated by strong coupling between excitons and cavity photons that follow the Bose statistics. They have been extensively investigated in the past decades for their unique properties, such as very light effective mass and the consequent high critical temperature for condensation [1]. It is well known that the occurrence of such a phase transition is accompanied by the onset of long range phase coherence [2]. In fact, since the unambiguous demonstration of polariton condensation its coherence properties have been investigated [3]. Moreover, the extension and eventually the decay of the spatial coherence can give useful information on the kind of transition the system undergoes [4, 5]. Recently, polariton condensates have attracted intense attention for their potential application in the field of quantum information, and also some properties suitable for such applications, such as Josephson oscillations and spin-switching, have been already demonstrated [6, 7]. Therefore the achievement of extended spatial and temporal coherence for the polariton condensate is crucial.

Here we study the spatial and temporal coherence properties of nonequilibrium polariton condensates under an optical parametric oscillator (OPO) regime. The OPO is a third order non-linear phenomenon, arising from interactions between the excitonic components of the polaritons. It takes place when two pump polaritons at the inflection point of the lower polariton branch (LPB), scatter efficiently into a signal and an idler polariton [8, 9]. Spatial coherence properties have been studied theoretically for the OPO condensate by Carusotto and Ciuti [10]. They investigated numerically the first order coherence function ($g^{(1)}$) for a finite condensate paying special attention to its behaviour across the OPO parametric threshold (E_{Th}) [11]. It is found that, for excitation frequencies ω_p below that of the threshold, $g^{(1)}$ has a finite correlation length. Increasing ω_p in order to approach E_{Th} , maintaining fixed the pump angle

(and therefore the wavevector), they predict the build up of macroscopic phase coherence extending over the entire condensate [10]. In this regime the spatial fluctuations are negligible, so the temporal coherence properties should be captured by the theory of Whittaker and Eastham [12]. In this theory the temporal coherence is limited by fluctuations in the particle number, which due to the polariton-polariton interactions imply a broadening of the emission [13–15]. This broadening mechanism, however, could be suppressed by motional narrowing [12], so that for appropriate pump powers and condensate areas very long coherence times could be obtained. Despite the fact that long coherence times and extended spatial coherence have been predicted theoretically for the OPO configuration, only now our experiments confirm such predictions.

One of the main factors that limits the coherence is the quality of the cavity: the presence of defects creates a disorder potential that traps the condensate, potentially leading to multi-mode and inhomogeneous states [16]. Another, very important, detrimental effect is caused by the fluctuations of the excitation laser that hinder the attainment of the intrinsic coherence of the condensate. Many studies [3, 16, 17] have been performed by non-resonantly pumping the microcavity, in such cases the resulting distribution of the population at the bottom of the lower polariton branch is subjected to fluctuations due to the reservoir of particles at the bottleneck. These fluctuations, broadening the distribution of polaritons in energy and momentum space, translate, according to the Wiener-Khinchin identity [18, 19], into a faster decay of the temporal and spatial coherence. Although analogous broadening by the fluctuating population of pump polaritons can occur in the OPO [15], the threshold pump density (P_{Th}) for a resonant-gain process is much lower than that for non-resonant gain and the effect of the reservoir polaritons is less important. Thus the coherence exhibited by an OPO polariton-condensate is expected to

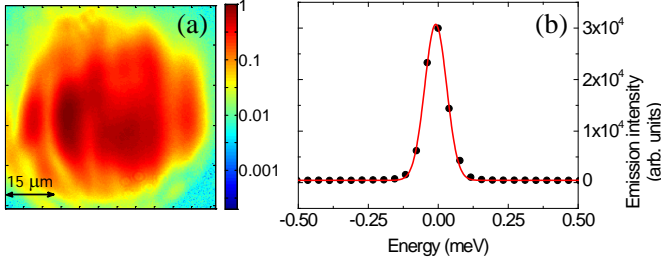


FIG. 1: (color online) (a) Typical real space emission intensity (color scale) from the signal condensate generated by parametric scattering, at detuning $\delta \sim -3.8$ meV and pump power $P = 10$ kW/cm². (b) Corresponding energy spectrum of the signal emission (spectrometer limited).

decay over much longer times and larger distances than the one produced by non-resonant techniques. To avoid that the pump fluctuations conceal the genuine coherence properties of the condensate we use a CW monomode laser with a very narrow bandwidth of 75 kHz to excite the sample, which is a high-quality λ -microcavity grown by molecular beam epitaxy, composed by 16 periods of $\lambda/4$ AlAs/Al_{0.15}Ga_{0.85}As on its top and 25 periods on its bottom. A single 10 nm GaAs/Al_{0.3}Ga_{0.7}As QW is placed at the antinode of the cavity. The Rabi splitting is 4.2 meV and we select a detuning of $\delta \sim -3.8$ meV. In order to investigate the spatial coherence properties of a true 2D condensate we create one with a diameter of ~ 40 μm, by pumping resonantly the LPB close to its inflection point, with a Gaussian laser beam, maintaining the temperature of the sample at 10 K. We adopt the experimental conditions to drive the sample into the OPO regime at the parametric threshold, E_{Th} , and then we follow a similar approach to that in [10], i.e., we decrease the pump frequency ω_p keeping fixed the angle of incidence. Typical results for the OPO signal are shown in Fig. 1. They reveal a bright and rather homogeneous emission in real space (Fig. 1(a)), with a spectrometer-limited emission spectrum (Fig. 1(b)).

Therefore, to evaluate the coherence of the condensate, since its true energy linewidth is masked by our spectrometer, the signal emission is analyzed with the help of a Mach-Zehnder interferometer. On one of the arms of the interferometer a retro reflector mirror is mounted, which flips the image of the condensate in a centrosymmetric way. Recombining, at the output of the interferometer, the image of the condensate with its flipped image allows evaluating the coherence of each point (x, y) with respect to its opposite counterpart $(-x, -y)$. In this way we access to the first order coherence function $g^{(1)}[(x, y), (-x, -y)] = \frac{\langle E^*(x, y)E(-x, -y) \rangle}{\langle E^*(x, y) \rangle \langle E(-x, -y) \rangle}$ [2, 3]. To extract the coherence of the condensate the retroreflector mirror is moved, using a piezo translation

stage, by a few wavelengths around the zero delay position, adding a short delay Δt to the retroreflector arm with respect to the other arm. For each measurement the intensities of the two arms were acquired and used in the following normalized expression of the interference pattern: $I_{Norm}[(x, y), (-x, -y)] = \frac{I_{Tot} - I_{RR} - I_M}{2\sqrt{I_{RR}I_M}} = g^{(1)}[(x, y), (-x, -y)] \sin(\Delta t + \varphi_0)$ where I_{Tot} is the total intensity of the interference pattern, I_{RR} (I_M) is the intensity recorded from the arm with the retroreflector (single mirror), and φ_0 is the initial phase of the condensate. For each point of the condensate a sinusoidal evolution of the normalized interference as a function of the delay Δt is recorded. The first order coherence of each point of the condensate is extracted with the help of the previous formula; and repeating this procedure for each point we can reconstruct the entire coherence map.

At E_{Th} , the interference pattern extends along the entire condensate (Fig. 2(a)) and the corresponding coherence map (Fig. 2(b)) reveals that a constant phase is developed through its entire area, validating the theoretical predictions [10]. This is shown in detail in Fig. 2(c), which depicts the coherence of a horizontal profile (0.26 μm width) taken at the center of the map. The fact that the degree of coherence is almost flat along the entire condensate demonstrates that only a single coherent mode is formed, in contrast with what has been found in previous works [16, 20] where disorder of the sample caused the development of several modes. The extension of the coherence here is ~ 40 μm, the largest reported until now in 2D microcavity polaritons, to the best of our knowledge; only in 1D microcavities, for propagating condensates, a larger spatial coherence has been shown [21]. It is worth to note that in an OPO process, the coherence properties of the signal are not inherited from the excitation laser [22]. Although the theory of an infinite 2D condensate predicts the absence of long-range order, and a power-law decay of the spatial coherence [4], in a finite dimension system such as ours, determined by the pump laser size, a constant coherence of the condensate can be obtained [3].

By decreasing the pump energy by $\delta E = 0.01$ meV towards lower energies, leaving fixed the pump angle, the phase matching conditions for the OPO are not fulfilled anymore, although an emission is obtained at the bottom of the LPB. The interferometric analysis of the emission from the cloud of non-condensed polaritons gives an interference pattern only at the center of the emission (Fig. 2(d)). The corresponding coherence map (Fig. 2(e)) reveals a ~ 7 μm coherence length, of a few polariton de Broglie wavelengths, extracted by fitting the profile of the spatial decay (Fig. 2(f)). These results demonstrate the predictions of Ref. [10] about the coherence of polariton emission below and at the condensation threshold.

Measuring $g^{(1)}$ at a given point for different time delays τ permits the evaluation of the signal temporal coherence

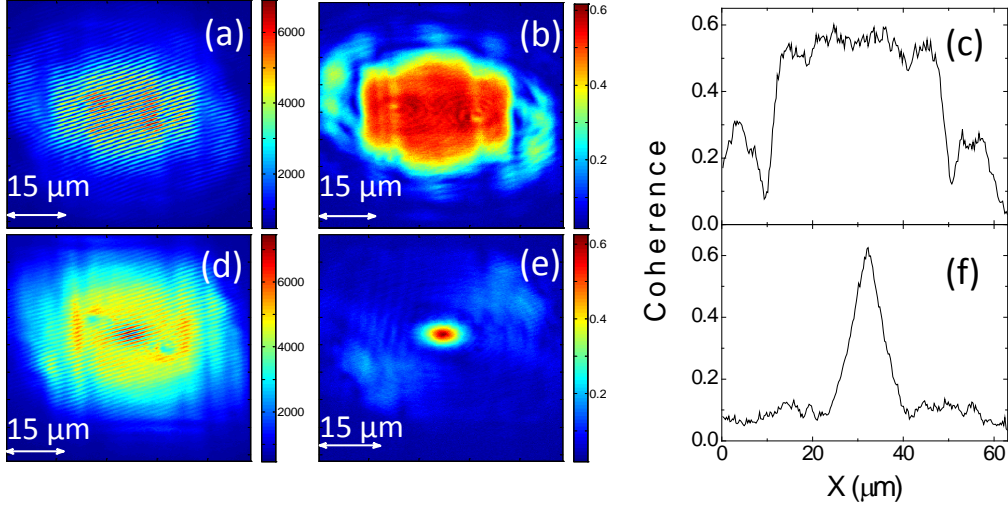


FIG. 2: (color online) Interference pattern (a/d) and corresponding coherence (b/e) of the condensate generated at/below the parametric threshold, E_{Th} , corresponding to a pump energy $E_1 = 1552.57$ meV / $E_2 = 1552.56$ meV and power density $P = 15.8$ kW/cm². Horizontal profiles at the center (c/f) showing the constant coherence along the entire condensate for E_1 , and an exponentially decaying one for E_2 .

$g^{(1)}(\tau) = \frac{\langle E^*(t)E(t+\tau) \rangle}{\langle E^*(t) \rangle \langle E(t) \rangle}$. In our case we obtain $g^{(1)}$ for different values of the condensate area A and pump power density P , as shown in Fig. 3. We obtain a predominantly exponential decay for $g^{(1)}(\tau)$, with a coherence time reaching $T_c = (3.2 \pm 0.8)$ ns. This is much longer than the largest value reported in the literature until now for the same material system [15].

Despite the lack of spatial fluctuations, the coherence time of our condensate, though long, is clearly finite. In general, such a finite correlation time in a state with perfect spatial order is caused by finite-size fluctuations [23], and reflects the absence of true phase transitions in finite systems. A well-known example is the Schawlow-Townes form for the coherence time of a single laser mode with an average of \bar{N} photons, $T_c \propto \bar{N}$. Since $\bar{N} \propto A$ when the control parameter P is constant, the Schawlow-Townes result implies the scaling form $T_c \propto A$. However, as shown in detail below, this scaling law is violated by more than an order of magnitude for our condensate, and our results cannot be understood as a straightforward effect of increasing the particle number. Our results may be interpreted in terms of the theoretical model of Ref. [12], in which the linewidth arises from fluctuations in the number of particles. Due to the interactions, such number fluctuations imply energetic fluctuations of the emission, leading to a broadened line or a decay of $g^{(1)}(\tau)$. This is captured by Kubo's result

$$|g^{(1)}(\tau)| = \exp \left[-\frac{2\tau_r^2}{\tau_c^2} \left(e^{-\tau/\tau_r} + \frac{\tau}{\tau_r} - 1 \right) \right], \quad (1)$$

for the emission from a transition whose energy fluctuates, where τ_c is determined by the width of the (generi-

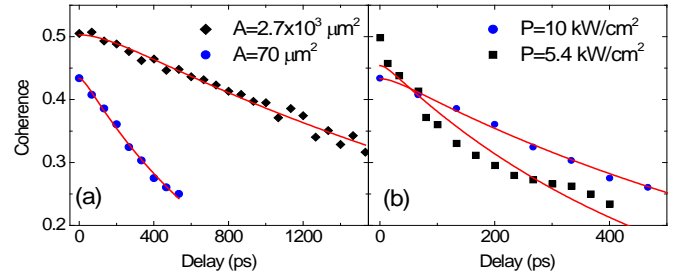


FIG. 3: (color online) (a) Temporal coherence decay above parametric threshold for condensate areas $A = 70 \mu\text{m}^2$ (blue dots) and $2.7 \times 10^3 \mu\text{m}^2$ (black diamonds) at a pump power density $P = 10$ kW/cm²; (b) Temporal coherence decay for $P = 5.4$ kW/cm² (black squares) and 10 kW/cm² (blue dots) at $A = 70 \mu\text{m}^2$. Lines are fits to Eq.(1).

cally Gaussian) distribution of the energy level and τ_r is the characteristic timescale on which the fluctuations occur. If $\tau_r \gg \tau_c$ the fluctuations are slow and the emission reflects the energy distribution of the level, so that the coherence decays on timescale τ_c . If however $\tau_r \ll \tau_c$ a motional narrowing effect leads to an exponential decay, with a much longer coherence time $T_c = \tau_c^2/2\tau_r$.

To elucidate the effects of number fluctuations, we fit the data in Fig. 3 to the Kubo formula (1). Pumping with the same power density $P = 10$ kW/cm², the condensate with a larger area $A = 2.7 \times 10^3 \mu\text{m}^2$ presents $\tau_c = (0.97 \pm 0.11)$ ns and $\tau_r = (0.15 \pm 0.02)$ ns fulfilling the inequality for the motional narrowing regime and giv-

ing a coherence time $T_c = (3.2 \pm 0.8)$ ns. With a smaller area $A = 70 \mu\text{m}^2$, we find a decrease of both parameters $\tau_c = (0.20 \pm 0.08)$ ns and $\tau_r = (0.023 \pm 0.007)$ ns, still indicating the system being in the motional narrowing regime but giving a shorter $T_c = (0.9 \pm 0.4)$ ns. As well as moving to larger areas, another way to have longer decay times suggested by [12] is to increase the pump power. The results shown in Fig. 3(b), corresponding to two different powers keeping now the size of the condensate constant, also demonstrate that increasing the power a longer coherence time is achieved. In fact the coherence time for a condensate of relatively small area $A = 70 \mu\text{m}^2$ goes from $T_c = (0.5 \pm 0.3)$ ns at $P = 5.4 \text{ kW/cm}^2$, to $T_c = (0.9 \pm 0.4)$ ns at $P = 10 \text{ kW/cm}^2$. We note that the coherence decay of the smallest condensate at lowest power, black squares in Fig. 3(b), has considerable structure and does not follow the Kubo form, suggesting that this condensate is not a single mode. From the Kubo fits to our data, for a given power, we can extract the scaling $\tau_c \propto A^{0.4 \pm 0.1}$. This is consistent with the dominant dephasing mechanism being the polariton-polariton interaction, suppressed by motional narrowing, in which case the Kubo formula should hold with $\tau_c \propto A^{0.5}$ [12]. Although this implies an increase in the coherence time, T_c , with area, our results show that this increase is restrained because τ_r also increases, so that motional narrowing becomes less effective.

The relatively small size of our system and the use of pump powers which are close to threshold, P_{Th} , are responsible for the observed increase of τ_r with area. In the thermodynamic limit, $A \rightarrow \infty$, the occupation, n , of a single mode with linear gain γ , linear loss γ_c and nonlinear gain Γ obeys the mean-field rate equation $dn/dt = (\gamma - \gamma_c - \Gamma n)n$ [24]. Linearizing we see that the damping time for number fluctuations is $\tau_r = 1/|\gamma - \gamma_c|$. This timescale is therefore independent of area when the rate equation is valid. However, at power threshold $\gamma = \gamma_c$ and the rate equation predicts a divergence in the relaxation time, which in the finite system must be cut off by fluctuations. Thus, in the threshold region, τ_r initially grows with area, as we observe, before eventually saturating. We obtain a similar behavior from numerical solutions to the dynamical model described in Ref. [12] in the threshold region. If we assume a polariton lifetime $\tau_0 = 3$ ps, then our results for $P = 10 \text{ kW/cm}^2$, $A = 70 \mu\text{m}^2$ imply the critical slowing factor $\tau_r/\tau_0 \approx 8$. Fitting this to the dynamical model at threshold gives a reasonable value for the gain saturation density, $\rho_s = n_s/A = 230/(70 \mu\text{m}^2)$. Since ρ_s is independent of A , we use this value in the model to predict $\tau_r = 140$ ps for our largest condensate, consistent with experiments.

For completeness we also studied the temporal coherence (not shown) under two further conditions: a) Exciting the LPB at the inflection point, for power densities slightly above but very close to P_{Th} , we find a Gaus-

sian decay of $g^{(1)}(\tau)$, as previously observed for condensates with non-resonant pumping [16]. This is consistent with the expected increase in τ_r approaching P_{Th} causing a crossover from the motional-narrowing to the static regime. b) Exciting far from the inflection point (no condensation achieved) we observe a very fast exponential decay of coherence on the range of the polariton lifetime.

In summary, we have investigated the spatial and temporal coherence properties of polariton condensates generated by parametric scattering. The spatial coherence was studied below and above the OPO condensation threshold, obtaining the largest value for the coherence length measured for 2D GaAs microcavity polaritons. Measurements of the temporal coherence reveal a predominantly exponential decay caused by polariton-polariton interactions in a motional narrowing regime. Although similar to the exponential decay associated with the Schawlow-Townes linewidth, the mechanism is completely different, and gives a different dependence of coherence time on condensate area. By constructing condensates of large area we are able to achieve long coherence times, which is a crucial step forward for exploiting polariton condensates in quantum and ultrafast devices.

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- [1] *Physics of Semiconductor Microcavities*, ed. B. Deveaud (Wiley-VCH, Berlin, 2007).
- [2] O. Penrose and L. Onsager, *Phys. Rev.* **104**, 576 (1956).
- [3] J. Kasprzak *et al.*, *Nature* **443**, 409 (2006).
- [4] L. Pitaevskii and S. Stringari, *Bose-Einstein condensation* (Oxford University Press, Oxford, 2003).
- [5] F. P. Laussy *et al.*, *Phys. Rev. Lett.* **93**, 016402 (2004).
- [6] K. G. Lagoudakis *et al.*, *Phys. Rev. Lett.* **105**, 120403 (2010).
- [7] A. Amo *et al.*, *Nature Photonics* **4**, 361 (2010).
- [8] R. M. Stevenson *et al.*, *Phys. Rev. Lett.* **85**, 3680 (2000).
- [9] J. J. Baumberg *et al.* *Phys. Rev. B* **62**, R16247 (2000).
- [10] I. Carusotto and C. Ciuti, *Phys. Rev. B* **72**, 125335 (2005).
- [11] In this work we consider two kinds of threshold, associated with tuning the pump energy and power. They are denoted E_{Th} and P_{Th} respectively.
- [12] D. M. Whittaker and P. R. Eastham, *EPL* **87**, 27002 (2009).
- [13] F. Tassone and Y. Yamamoto, *Phys. Rev. A* **62**, 063809 (2000).
- [14] D. Porras and C. Tejedor, *Phys. Rev. B* **67**, 161310(R) (2003).

- [15] D. N. Krizhanovskii *et al.*, Phys. Rev. Lett. **97**, 097402 (2006).
- [16] D. N. Krizhanovskii *et al.*, Phys. Rev. B **80**, 045317 (2009).
- [17] H. Deng *et al.*, Phys. Rev. Lett. **99**, 126403 (2007).
- [18] H. P. Baltes, Appl. Phys. **12**, 221 (1977).
- [19] M. Richard, M. Wouters and L.S. Dang, in *Optical Generation and Control of Quantum Coherence in Semiconductor Nanostructures*, NanoScience and Technology **146**, Chap.11, eds. G. Slavcheva and P. Roussignol (Springer-Verlag Berlin 2010).
- [20] A. P. D. Love *et al.*, Phys. Rev. Lett. **101**, 067404 (2008).
- [21] E. Wertz *et al.*, Nature Phys. **6**, 860 (2010).
- [22] M. Wouters and I. Carusotto, Phys. Rev. A **76**, 043807 (2007).
- [23] P. R. Eastham and P. B. Littlewood, Phys. Rev. B **73**, 085306 (2006).
- [24] P.R. Eastham, Phys. Rev. B **78**, 035319 (2008).